

# Synthesis of neutron-rich transuranic nuclei in fissile spallation targets

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## Abstract

A possibility of synthesizing neutron-rich super-heavy elements in spallation targets of Accelerator Driven Systems (ADS) is considered. A dedicated software called Nuclide Composition Dynamics (NuCoD) was developed to model the evolution of isotope composition in the targets during a long-time irradiation by intense proton and deuteron beams. Simulation results show that transuranic elements up to  $^{249}\text{Bk}$  can be produced in multiple neutron capture reactions in macroscopic quantities. However, the neutron flux achievable in a spallation target is still insufficient to overcome the so-called fermium gap. Further optimization of the target design, in particular, by including moderating material and covering it by a reflector will turn ADS into an alternative source of transuranic elements in addition to nuclear fission reactors.

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## 1. Introduction

Neutrons propagating in a medium induce different types of nuclear reactions depending on their energy. Apart of the elastic scattering, the main reaction types for low-energy neutrons are fission and neutron capture, which dominate, respectively, at higher and lower energies with the boarder line around 1 MeV. Generally, the average number of neutrons captured by a nucleus  $A$  during a time interval of  $\Delta t$  is given by the formula:

$$\Delta N = f \sigma_{nA} \Delta t , \quad (1)$$

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where  $\sigma_{nA} = (1 \div 3) \text{ b}$  is the cross section of  $(n, \gamma)$  reaction on heavy nuclei and  $f = \langle v \frac{dn}{dv} \rangle$  is the neutron flux averaged over the velocity distribution.

The possibility to capture many neutrons in nuclear explosions was considered already in 60s and 70s, see e.g. ref. [1], and recently in refs. [2, 3]. In this case the neutron flux is about  $3 \cdot 10^{30} \frac{\text{n}}{\text{s} \cdot \text{cm}^2}$ , and the explosion time  $\Delta t \simeq 1 \text{ } \mu\text{s}$ , therefore:

$$\Delta N_{\text{expl}} = f \Delta t \sigma = 3 \cdot 10^{30} \frac{\text{n}}{\text{s} \cdot \text{cm}^2} \cdot 10^{-6} \text{s} \cdot 10^{-24} \text{cm}^2 \simeq 3,$$

assuming that  $\sigma_{nA} \simeq 1 \text{ b}$ . As shown in [3], macroscopic quantities of superheavy (SH) elements located on the Island of Stability can be produced in multiple nuclear explosions.

In ordinary nuclear fission reactors the average neutron flux is typically below  $10^{15} \text{ n}/(\text{s} \cdot \text{cm}^2)$ , and the nuclei up to fermium ( $Z=100$ ) can be produced there [4]. In this paper we consider the possibility of producing neutron-rich transuranic (TRU) nuclei in Accelerator Driven Systems (ADS) with fissile spallation targets made of americium ( $Z=95$ ). In this case the average neutron flux could be as large as  $3 \cdot 10^{16} \text{ n}/(\text{s} \cdot \text{cm}^2)$  [5]. Since the  $(n, \gamma)$  reaction is only efficient at relatively low energies,  $E_n < 1 \text{ MeV}$ , the flux of such neutrons is about twice as low. However, the target can be irradiated by a proton or deuteron beam for a long time, e.g. for one year. Then taking  $\sigma_{nA} = 2 \text{ b}$ , one obtains:

$$\Delta N_{\text{ADS}} \simeq 3 \cdot 10^{16} \frac{\text{n}}{\text{s} \cdot \text{cm}^2} \cdot 3 \cdot 10^7 \text{s} \cdot 2 \cdot 10^{-24} \text{cm}^2 \simeq 2$$

The distribution of nuclides in the number of captured neutrons follows a Poisson distribution, providing that all other reactions are neglected, see, e.g. [2]. With a simple-minded correction for losses due to fission it can be written as:

$$P_n(\bar{n}) = \frac{\bar{n}^n e^{-\bar{n}}}{n!} (1 - q_{\text{fission}})^n \quad (2)$$

where  $q_{\text{fission}} \simeq \frac{1}{2}$  is the probability of fission in each step. With  $\bar{n} = 2$  and  $q_{\text{fission}} = \frac{1}{2}$  the probability of capture of 20 neutrons, to produce isotopes with atomic mass number  $A + 20$ , can be estimated as  $6 \cdot 10^{-20}$ . For a small Am target with its mass of about 40 kg the number of nuclei in the target is  $\sim 10^{26}$ , therefore,  $\sim 10^6$  nuclei with mass  $A+20$  can be synthesized after one year of operation.

Neutrons suitable for the production of super-heavy elements should have softer spectrum to increase the efficiency of neutron capture reactions. Therefore, such targets should include some moderating material to slow down neutrons. However, too intensive moderation of neutrons would suppress the neutron breeding in fission reactions. A special investigation is required to find a compromise solution. In this paper we model an Am target equipped with a Be reflector by performing detailed Monte Carlo simulations of nuclear reactions and using nuclear data files with realistic neutron interaction cross sections.

## 2. Modelling of fissile spallation targets

### 2.1. MCADS — Monte Carlo model for Accelerator Driven Systems

Simulations in this work are performed with the Monte Carlo model for Accelerator Driven Systems (MCADS), which is based on the Geant4 toolkit (version 9.4 with patch 01) [6, 7, 8]. This model was used for modeling neutron production and transport in spallation targets made of tungsten, uranium and americium [9, 5]. MCADS is capable to visualize target volumes as well as histories of primary protons and all secondary particles. It has flexible scoring techniques to calculate neutron flux, heat deposition inside the target and leakage of particles from the target. MCADS makes possible to employ several cascade models for the fast initial stage of pA and dA interactions, which are combined with evaporation, multi-fragmentation and Fermi break-up models for the slow de-excitation stage of thermalized residual nuclei.

MCADS was validated with three different cascade models: Bertini Cascade and Binary Cascade coupled with their standard fission-evaporation codes and Intra-Nuclear Cascade Liège (INCL) coupled with the ABLA model. Calculations were performed for thin and thick targets made of tungsten and uranium [9]. For thin  $^{238}\text{U}$  targets irradiated by 27, 63 and 1000 MeV protons we have analyzed data on the fission cross sections, neutron multiplicity and mass distribution of fission products, and the best agreement was obtained for calculations involving INCL/ABLA. In particular, as demonstrated in ref. [9] MCADS results obtained with INCL/ABLA model are in good agreement with experimental data available for extended tungsten and uranium targets irradiated by protons in the energy range of 400 – 1470 MeV.

Several extensions of the Geant4 source code and additional nuclear data [10] are needed for the description of proton- and neutron-induced reactions and

elastic scattering on TRU nuclei [11]. MCADS with the modified Geant4 was validated with  $^{241}\text{Am}$  and  $^{243}\text{Am}$  thin targets. A good agreement with proton- and neutron-induced fission cross sections, fission neutron spectra, neutron multiplicities, fragment mass distributions and neutron capture cross sections measured in experiments was demonstrated [12].

## 2.2. Criticality studies

When dealing with fissile materials one must control the criticality of the target. This can be done by calculating the neutron multiplication factor  $k = \langle N_i/N_{i-1} \rangle$ , which is defined as the average ratio between the numbers of neutrons,  $N_{i-1}$  and  $N_i$ , in two subsequent generations  $i-1$  and  $i$ . Obviously, the target mass should not exceed the so-called critical mass which corresponds to the condition  $k = 1$ . The calculated number of neutrons in the first 50 neutron generations developing in a cylindrical  $^{241}\text{Am}$  target following an impact of a beam proton is shown in Figure 1. The results are given for a set of targets of the same length (150 mm), but different radii in the range from 40 to 110 mm. As one can see,  $k \simeq 1$  in the target with the radius of 106 mm, and the number of neutrons in subsequent generations remains constant. A supercritical regime ( $k > 1$ ) is reached for larger targets where the number of neutrons grows from one generation to another.

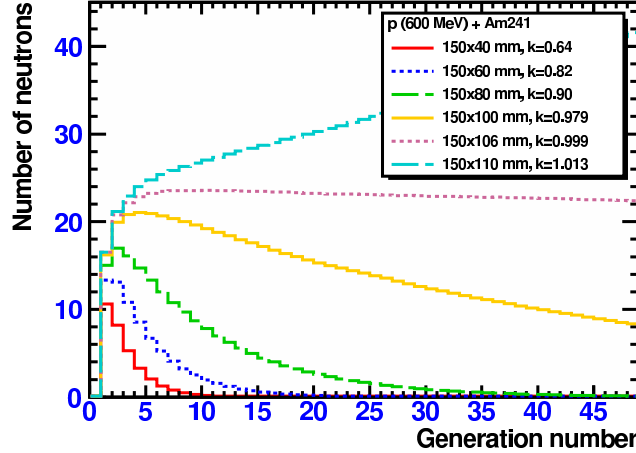


Figure 1: Number of neutrons  $N_i$  in  $^{241}\text{Am}$  targets of 15 cm length and radii of 40, 60, 80, 100, 106 and 110 mm as a function of neutron generation number  $i$ .

The main safety requirement to a spallation target made of a fissile material is that it is deeply subcritical and operates in a safe mode. Cylindrical Am targets, which are characterized by the neutron multiplication factor  $k \leq 0.6$ , are considered below.

### **3. Changes in isotope composition of spallation targets during irradiation**

#### *3.1. NuCoD – Nuclide Composition Dynamics*

A significant amount of nuclides can be produced and accumulated in a spallation target during its long-term irradiation. Moreover, nuclides of interest can be produced specifically in nuclear reactions on those nuclides, which were absent in the original target material before the beginning of irradiation. In this respect the MCADS model has a disadvantage of neglecting reactions induced on new nuclides produced in the target during irradiation. A burn-up of the target material is also neglected. This stems from a restriction of the Geant4 toolkit which is incapable of changing the definitions of materials used to build geometry objects until a required number of events is obtained, i.e. during a simulation run. This restriction does not impair the accuracy of MCADS results for a short-term irradiations, when such changes can be neglected.

The Nuclide Composition Dynamics (NuCoD) code was developed on the basis of MCADS for modeling long-term irradiation of spallation targets taking into account the above-described changes of their nuclide composition during the irradiation period. Nuclides produced in spallation targets can be transmuted into other nuclei in reactions induced by beam projectiles and various secondary particles. Furthermore, unstable isotopes undergo radioactive decays. The algorithm of modeling consists of several steps:

1. Monte Carlo simulation with a target of a given composition to calculate the yields of products of spallation, fission and neutron capture reactions;
2. calculation of changes of such yields in time due to respective radioactive decays;
3. update of the target composition and preparing input for the next Monte Carlo simulation.

Following this simulation scheme the Geant4-based code MCADS described above in Section 2.1 is used for Monte Carlo simulations with targets of constant

nuclide composition. MCADS is also used to verify that each target composition is subcritical. In turn, NuCoD uses the yields of newly produced and burned-up nuclides calculated by MCADS at a previous step to estimate the isotope abundances in the target for a given beam intensity and exposure time. The key feature of MCADS consists in the ability to simulate targets containing minor actinides (MA). This makes possible to calculate the yields of TRU elements produced in a long chain of nuclear reactions and radioactive decays in targets containing MA by using NuCoD.

It is not reasonable to run MCADS for the targets containing all the isotopes produced at previous steps. Firstly, it is too expensive in terms of CPU time, as simulations with multicomponent materials require more computational steps. Secondly, many isotopes in the target do not influence significantly the yields of the isotopes of our interest and thus may be omitted. Thirdly, the abundances of some isotopes may be too low. As a result, the number of reactions generated in an MCADS run on such rare nuclei may be insufficient for reliable estimations of the yields of respective reaction products.

In order to solve all these problems, a special technique was implemented. Instead of modeling a target containing all the isotopes with their actual abundances, a set of special targets is considered at each simulation step  $n$ . The first one, called the base target, contains only the most abundant isotopes. Then, separate modified targets, referred to as isotope-enriched targets, are constructed for each isotope  $(A_i, Z_i)$  with its abundance  $a(A_i, Z_i, n)$  less than  $a_{th1} = 0.005$ , but which affects significantly the final yields of isotopes we are interested in. These modified targets are constructed as follows:

- The base target contains only isotopes with abundances exceeding a threshold value  $a_{th0}$  (by default  $a_{th0} = 0.001$ ). Additionally, the isotopes, for which the isotope-enriched targets are built, are excluded from the base target.
- In the  $i$ -th isotope-enriched target the concentration of the isotope  $i$  is enhanced artificially. Its abundance is set to  $a_{th1} = 0.005$  and the corresponding scale factor  $k_i(n)$  is stored.

Running MCADS with these targets makes possible to calculate the amounts of newly produced and consumed isotopes at each time step  $\Delta t$ .  $\Delta \tilde{a}(A, Z, n)$  is defined as the abundance variation of an isotope with mass  $A$  and charge  $Z$

calculated with MCADS for the base target during the step  $n$ . Analogously,  $\Delta a_i(A, Z, n)$  is calculated for each isotope-enriched target. Finally, the abundance of the isotope with mass  $A$  and charge  $Z$  at the beginning of the step  $(n + 1)$  is calculated as:

$$a(A, Z, n+1) = a(A, Z, n) + \Delta \tilde{a}(A, Z, n) + \sum_i \frac{1}{k_i(n)} (\Delta a_i(A, Z, n) - \Delta \tilde{a}(A, Z, n)). \quad (3)$$

Finally, the evolution of the target composition due to decays is calculated using a newly developed recursive algorithm, which in principle can be considered as an extension of Bateman equations [13] for the case of a continuously irradiated target. This algorithm is described in Appendix A.

### 3.2. NuCoD validation

In order to validate NuCoD an irradiation of a lead target by protons as described in [14] was simulated. In the experiment [14] a target with the diameter of 20 cm and length of 60 cm was irradiated by a 1 GeV proton beam with a current of 0.3  $\mu$ A for 3 hours. The measurements of  $\gamma$ -spectra started 7 hours after the termination of the irradiation and lasted next 8 days. In Figure 2 the yields of spallation products measured at the depth of 6 cm are compared with results of NuCoD simulation. Results of calculations obtained only with MCADS, i.e. neglecting decays of spallation products, are also shown in Figure 2 together with the results of CASCADE/INPE code reported in ref. [15].

As seen from Figure 2, the results of both codes, CASCADE/INPE and MCADS, deviate from the measured yields. However, CASCADE/INPE results are much closer to the data compared to MCADS, which calculates prompt yields and neglects radioactive decays of spallation products. In contrast, NuCoD simulates isotope production in the target and also respective decay chains. This notably improves the agreement with the measured yields. This is especially important for the domain of lighter elements (from Pt to Lu), where their abundances are significantly increased due to decay products of parent nuclides.

## 4. Isotope production in a $\text{Am}(\text{OH})_3$ target

### 4.1. Geometry and composition of the target

The yields of heavy transuranic elements, up to fermium, were calculated with NuCoD in a cylindrical target irradiated by a 1 GeV deuteron beam with

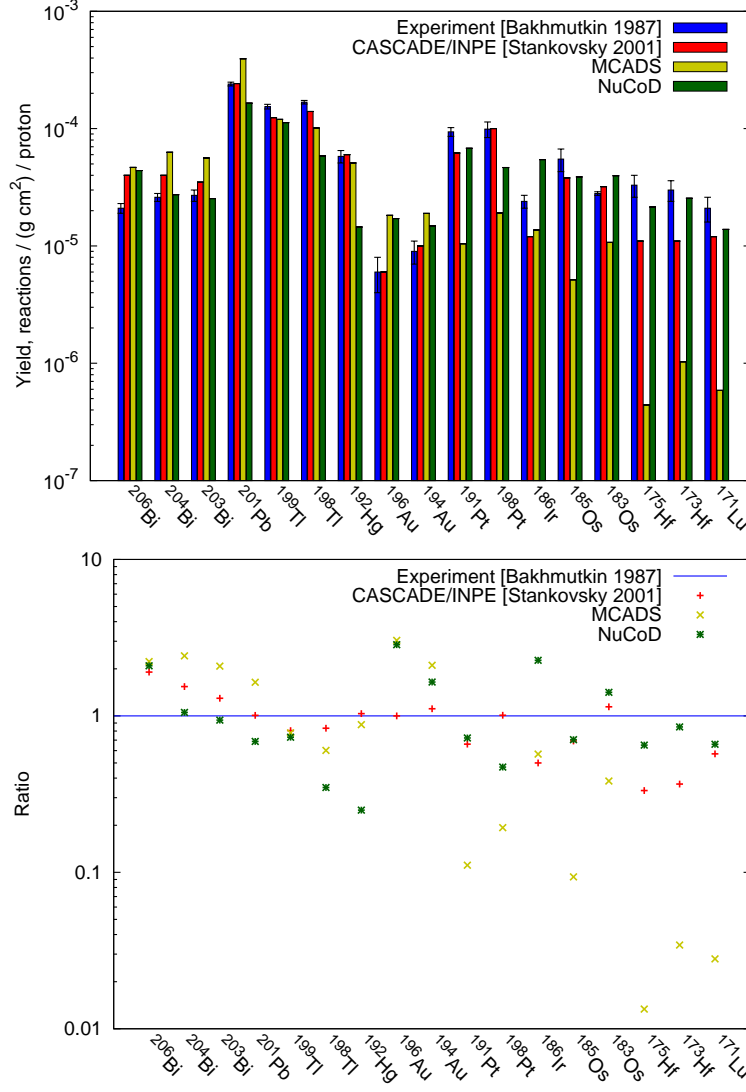


Figure 2: Yields of spallation products in the lead target irradiated by 1 GeV protons (top panel). Experimental data are from ref. [14]. Results of calculations with CASCADE/INPE [15], MCADS and NuCoD are presented by different colors explained in the legend. The calculated yields divided by the measured yields (bottom panel), see the legend for notations.

a current of 20 mA. The advantage of deuterons over protons with respect to neutron production in spallation targets was demonstrated in ref. [16]. This motivated our choice of projectiles in the present work. We consider a target of the 6 cm radius and 36 cm length made of  $\text{Am}(\text{OH})_3$ . Due to a lack of infor-



mation on the americium hydroxide properties its density was set to  $10 \text{ g/cm}^3$ . With this density of the target material the mass of the target was estimated as 40.7 kg.

Since the masses of neutrons and protons are almost equal, fast neutrons transfer to protons a half of their energy on average in their elastic scattering on hydrogen nuclei. Thus hydrogen containing in  $\text{Am}(\text{OH})_3$  moderates neutrons very efficiently. The probability of capture increases with the decrease of neutron energy and the production rate of neutron-rich elements also increases. In order to prevent, at least partially, leakage of neutrons, it was assumed that the target was covered by a 12 cm thick beryllium layer as shown in Figure 3. Since beryllium has a low neutron capture cross section, this material is widely used for reflecting neutrons in nuclear reactors.

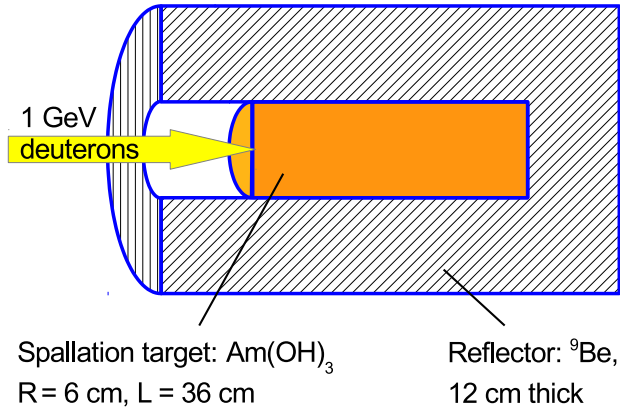


Figure 3: Layout of a spallation target for production of SH nuclei.

The isotope ratio of americium was taken approximately as in spent nuclear fuel,  $^{241}\text{Am} : ^{243}\text{Am} = 3 : 2$ . Since the probability to capture a certain number of neutrons drops rapidly with the number of required capture events, the presence of  $^{243}\text{Am}$  in the target enhances the production of neutron-rich SH nuclei due to two additional neutrons containing in  $^{243}\text{Am}$  with respect to  $^{241}\text{Am}$ . According to our Monte Carlo simulation results, the average number of neutrons captured by  $^{243}\text{Am}$  or heavier nuclides during 6 months of irradiation is about  $\bar{n}(0.5\text{y}) = 0.6$  and  $\bar{n}(1\text{y}) = 1.2$  during one year of irradiation. This is a more accurate result compared with the estimation given above in Section 1.

#### 4.2. Dynamical nuclear charts

Below we present our results obtained with the time step of 5 days between the target composition updates. The maps of isotopes contained in the target after 5, 30, 90 and 180 days of irradiation are presented in Figure 4. The first map represents the isotopes produced before the first target composition update, i.e. a direct output of MCADS simulation corrected for the radioactive decays. Therefore, only products of (d,Am), (p,Am) and (n,Am) reactions and their decay products are present on this map.

The main path to produce TRU elements (starting from  $^{243}\text{Am}$ ) is shown by thick arrows extending up to  $^{249}\text{Bk}$ .  $^{244}\text{Am}$  is produced in neutron capture reaction, and it undergoes  $\beta^-$ -decay with the half-life time of  $t_{1/2} = 10$  hours. A resulting long-lived isotope  $^{244}\text{Cm}$  ( $t_{1/2} = 18$  years) can capture more neutrons. The next four isotopes of Cm have half-life times of several thousands years and longer and can be accumulated in the target after corresponding number of  $(n, \gamma)$  reactions. The isotope  $^{249}\text{Cm}$  has the half-life time of 64 minutes and decays into a relatively long-lived  $^{249}\text{Bk}$  ( $t_{1/2} = 330$  days) which we are interested in. Heavier isotopes are produced in similar processes depicted by thin arrows in Figure 4 which extend up to fermium. The isotopes  $^{254-256}\text{Fm}$  have half-life times of 2.5–20 hours and undergo  $\alpha$ -decay or spontaneous fission and drop out from the chain of subsequent neutron capture reactions. That is why this specific domain in the map of isotopes is called the fermium gap. Due to its presence a further move to heavier isotopes is practically impossible. Indeed, as seen from Figure 4, the abundance of Fm isotopes in the considered spallation targets is extremely low:  $\sim 10^{-17}$ .

The production of  $^{250}\text{Cm}$  and  $^{254}\text{Es}$  is suppressed, respectively, by a prompt  $\beta^-$ -decay of  $^{249}\text{Cm}$  and the  $\alpha$ -decay of  $^{253}\text{Es}$ . However, the situation may change in the case of a pulse-mode accelerator with a short pulse duration of  $\sim 1 \mu\text{s}$  and very intense bunches of protons.

#### 4.3. Abundances of produced nuclides

The time evolution of the abundances of  $^{243}\text{Am}$ ,  $^{244,246}\text{Cm}$ ,  $^{249}\text{Bk}$ ,  $^{251}\text{Cf}$ ,  $^{254}\text{Es}$  and  $^{257}\text{Es}/^{257}\text{Fm}$  is shown in Figure 5. As one can see, the abundance of  $^{244}\text{Cm}$  raises rapidly at the beginning and after  $\sim 135$  days exceeds the  $^{243}\text{Am}$  abundance. In order to produce  $^{246}\text{Cm}$  three neutrons must be captured by a single nucleus. This means that  $^{246}\text{Cm}$  can be obtained only after three

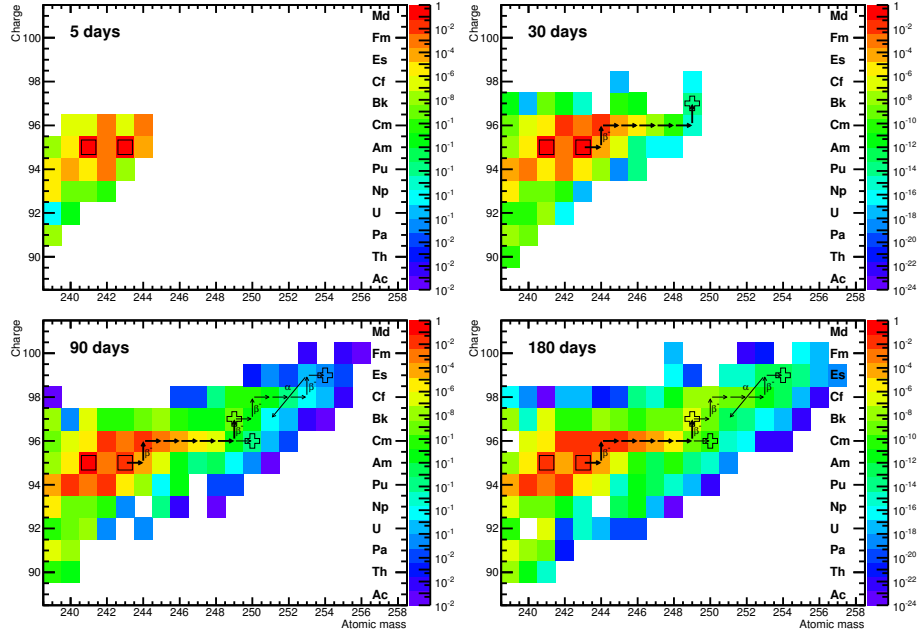


Figure 4: Calculated maps of isotope abundance after 5, 30, 90 and 180 days of irradiation. The initial isotopes,  $^{241}\text{Am}$  and  $^{243}\text{Am}$ , are marked with the squares, the crosses indicate  $^{249}\text{Bk}$ ,  $^{250}\text{Cm}$  and  $^{254}\text{Es}$ .

simulation steps of NuCoD, i.e. after 15 days. This deficiency of the simulation method is noticeable only at the first steps and much less affects the results in further steps. As seen from Figure 5, the amount of  $^{249}\text{Bk}$  reaches the level of 10 mg after 150 days.

The abundance of selected long-lived isotopes after 2, 4 and 6 months of irradiation are presented in Table 1. One can see that the amount of  $^{249}\text{Bk}$  exceeds the level of several milligrams, which is sufficient to fabricate a fusion target [17] to be used in experiments on production of SHE elements in nucleus-nucleus collisions. Due to the  $\beta^-$ -decay of  $^{249}\text{Bk}$  ( $t_{1/2} = 320$  days)  $^{249}\text{Cf}$  can also reach this level after some cooling time after the end of irradiation by deuterons.

The yields of neutron-rich elements rapidly increase if the irradiation time becomes longer. If the irradiation time is prolonged by a factor of 2, i.e. from six months to one year, the corresponding amounts of neutron-rich elements will

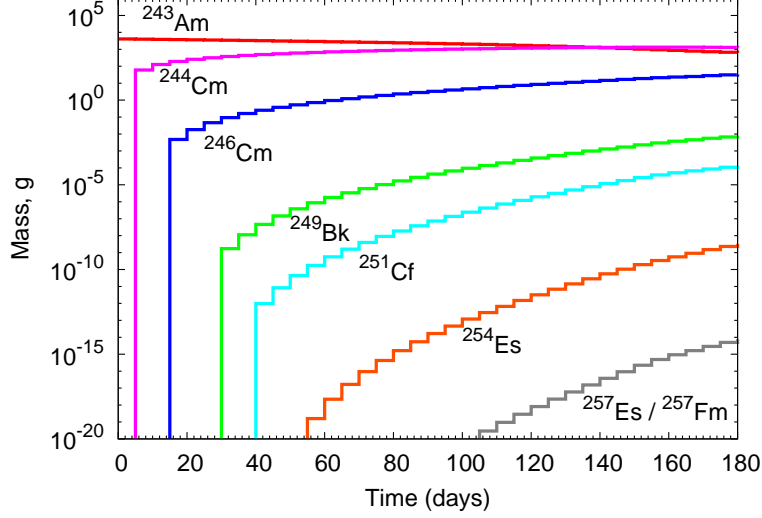


Figure 5: Time evolution of the amount of  $^{243}\text{Am}$ ,  $^{244}\text{Cm}$ ,  $^{246}\text{Cm}$ ,  $^{249}\text{Bk}$ ,  $^{251}\text{Cf}$ ,  $^{254}\text{Es}$  and  $^{257}\text{Es}/^{257}\text{Fm}$  calculated with NuCoD for the  $\text{Am}(\text{OH})_3$  target irradiated by 1 GeV deuteron beam.

Table 1: Abundance of  $^{249}\text{Bk}$ ,  $^{249,251}\text{Cf}$ ,  $^{250}\text{Cm}$ ,  $^{254}\text{Es}$  and  $^{257}\text{Es}/^{257}\text{Fm}$  after 2, 4 and 6 months of irradiation by a 1 GeV deuteron beam with a current of 20 mA.

Isotope	Mass (mg)		
	2 months	4 months	6 months
$^{249}\text{Bk}$	$7.5 \cdot 10^{-3}$	1.6	32
$^{249}\text{Cf}$	$1.0 \cdot 10^{-4}$	0.042	1.2
$^{250}\text{Cm}$	$2 \cdot 10^{-7}$	$1.1 \cdot 10^{-4}$	$4.1 \cdot 10^{-3}$
$^{251}\text{Cf}$	$2 \cdot 10^{-6}$	$8.2 \cdot 10^{-3}$	0.55
$^{254}\text{Es}$	$10^{-14}$	$10^{-8}$	$1.4 \cdot 10^{-5}$
$^{257}\text{Es}/^{257}\text{Fm}$	—	$3 \cdot 10^{-15}$	$3 \cdot 10^{-11}$

increase by a factor of

$$F(n) = 2^n \frac{e^{-\bar{n}(1y)}}{e^{-\bar{n}(0.5y)}} = 0.55 \cdot 2^n$$

where  $n$  is the number of captured neutrons, see Eq. (2). For  $^{257}\text{Es}$   $n = 14$  and  $F(14) \simeq 10^4$ . According to the results of NuCoD calculations the mass of accumulated  $^{257}\text{Es}$  is  $3 \cdot 10^{-11}$  mg, after half a year of irradiation, see Table 1. However, it was assumed that  $^{257}\text{Es}$  does not decay, while in reality it has a half-life time of about 7 days and undergoes  $\beta^-$ -decay into  $^{257}\text{Fm}$ , which has the half-life time of 100 days. This means that calculated mass of  $^{257}\text{Es}$  actually

corresponds to  $^{257}\text{Fm}$ . Multiplying  $3 \cdot 10^{-11}$  mg by the factor of  $F(14) \simeq 10^4$  one can obtain that the mass of accumulated  $^{257}\text{Fm}$  should be about  $10^{-7}$  mg after one year of irradiation.

## 5. Conclusions

We have extended the MCADS model previously used for simulations of spallation targets irradiated by intensive proton and deuteron beams to account for changes of isotopic composition of the spallation target during a long-term irradiation. For this purpose a dedicated software called Nuclide Composition Dynamics (NuCoD) was developed on the basis of MCADS. It models spallation targets taking into account the time evolution of their composition due to primary and secondary nuclear interactions and radioactive decays including nuclear reactions on spallation products. NuCoD was validated against available experimental data [14] on nuclide composition in a Pb spallation target after its irradiation by a proton beam.

NuCoD was used to study the possibility to produce transuranic neutron-rich super-heavy nuclei in spallation targets. It was found that macroscopic quantities of isotopes can be produced up to  $^{249}\text{Bk}$ . Therefore, ADS can be considered as an alternative for reactors currently used for the TRU elements production. It was demonstrated that  $^{257}\text{Fm}$  can be produced at least in picogram level with the state-of-the-art accelerator technology. Moreover, the production rate of SH nuclei can be increased with farther improvements of the system. For example, if the neutron capture rate would be enhanced by a factor of 4, e.g. by adding moderator materials, the abundance of  $^{257}\text{Fm}$  would increase by 7 orders of magnitude. Therefore, future development of accelerator technology together with optimization of the target assembly can open a new opportunity for producing neutron-rich heavy and super-heavy elements in macroscopic quantities.

## Acknowledgments

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## Appendix A – Calculation of decay chains

The rate of the nuclide composition change  $v(A, Z, n)$  between the steps  $n + 1$  and  $n$  caused by primary and secondary particle-nucleus interactions can be calculated as:

$$v(A, Z, n) = (a(A, Z, n + 1) - a(A, Z, n)) \cdot I \cdot 6.241 \cdot 10^{15}, \quad (4)$$

where  $I$  is the beam current in mA, which is multiplied by the factor of  $6.241 \cdot 10^{15}$  to provide the beam current in particles per second. Below the arguments  $A$ ,  $Z$  and  $n$  are omitted for the sake of simplicity. Then the reduction of  $a_0$ , the abundance of an isotope in the target, due to its decay is defined by the formula:

$$\frac{da_0(t)}{dt} = v_0 - \frac{a_0(t)}{\tau_0}, \quad (5)$$

where the index “0” is referred to the parent isotope in a decay chain and  $\tau_0$  is its mean lifetime. The solution of Eq. (5) is:

$$\begin{aligned} a_0(t) &= b_0 + c_0^0 e^{-t/\tau_0} \\ b_0 &= v_0 \tau_0 \\ c_0^0 &= a_0(0) - v_0 \tau_0. \end{aligned} \quad (6)$$

The abundance variation of the direct product of the parent isotope with the abundance  $a_0$  is defined by the formula:

$$\frac{da_1(t)}{dt} = \frac{a_0(t)}{\tau_0} - \frac{a_1(t)}{\tau_1}, \quad (7)$$

where  $\tau_1$  is the mean lifetime of the daughter isotope. The solution of this equation is:

$$\begin{aligned} a_1(t) &= b_1 + c_1^0 e^{-t/\tau_0} + c_1^1 e^{-t/\tau_1} \\ b_1 &= v_0 \tau_1 \\ c_1^0 &= (a_0(0) - v_0 \tau_0) \frac{\tau_1}{\tau_0 - \tau_1} \\ c_1^1 &= -v_0 \tau_1 - (a_0(0) - v_0 \tau_0) \frac{\tau_1}{\tau_0 - \tau_1}. \end{aligned} \quad (8)$$

For the subsequent isotopes in the decay chain:

$$\frac{da_i(t)}{dt} = \frac{a_{i-1}(t)}{\tau_{i-1}} - \frac{a_i(t)}{\tau_i}. \quad (9)$$

The solutions can be obtained recurrently:

$$\begin{aligned} a_i(t) &= b_i + \sum_{k=0}^i c_i^k e^{-t/\tau_k} \\ b_i &= b_{i-1} \frac{\tau_i}{\tau_{i-1}} \\ c_i^k &= c_{i-1}^k \frac{\tau_i \tau_k}{\tau_{i-1}(\tau_k - \tau_i)}, \quad k \neq i \\ c_i^i &= -b_i - \sum_{k=0}^{i-1} c_i^k. \end{aligned} \quad (10)$$

A simple Python-code calculates abundance variation rates  $v(A, Z, n)$  for all the isotopes using the output of MCADS simulation with the base target and the isotope-enriched targets according to Eqs. (3) and (4). Then the code runs through all the isotopes, either existed in the primary target at the beginning of the step or with a non-zero value of  $v(A, Z, n)$ , and recalculates the corresponding abundance according to Eq. (6). For stable isotopes  $\tau = \infty$  is taken. After that, the abundance of all the subsequent decay products are calculated according to Eq. (10).

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